On the renewed growth of atmospheric methane

Alexander J. Turner\textsuperscript{1,*}

\textsuperscript{1}Harvard University

*aturner@fas.harvard.edu

Harvard University

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Why do we care about atmospheric methane?

1) Methane is a potent greenhouse gas
   - 2nd only to CO₂

2) Recent trends in atmospheric methane are not well understood

Emissions based radiative forcing [W m⁻²]

- CO₂
- CH₄
- O₃
- H₂O (strat)

IPCC (2013)

(MLO data c/o NOAA)
Global burden is balanced by the sources and sinks

**Sources**

- **Anthropogenic**
  - Biomass Burning (5%)
  - Landfills (11%)
  - Ruminants (13%)
  - Rice (4%)
  - Fossil Fuels (14%)

- **Natural**
  - Wetlands (32%)
  - Geological (8%)

**Sinks**

- Tropospheric OH (90%)

Isotopic Composition ($\delta^{13}C$; ‰)

- Pyrogenic
- Thermogenic
- Biogenic

Kirschke et al. (2013)
Global burden is sensitive to small changes

\[
[\text{CH}_4](t) = \tau E(t) - (\tau E(t) - [\text{CH}_4]_0) e^{-t/\tau}
\]

Lifetime = 9.4 yr

Emissions = 550 Tg/yr
Global burden is sensitive to small changes

3% increase in emissions could explain the renewed growth
**Spring 2016 was an exciting time for methane!**

**Turner et al., GRL (2016):**
- Find a 30% increase in US methane emissions since 2002
- Could account for more than 30% of renewed growth
- Did not attribute increase to a particular sector

**Schaefer et al., Science (2016):**
- Thermogenic emissions did not cause the renewed growth
- Renewed growth is due to biogenic sources
- Most likely source is Asian livestock
Previous top-down estimates of US methane emissions

- Prior EDGAR (Bottom-up)
- Wecht et al. SCIAMACHY (2004)
- This work GOSAT (2009 – 2011)

Turner et al., ACP (2015)
Top-down studies point to an increase in US methane, not seen in bottom-up estimates

Turner et al., GRL (2016)
What data do we have to evaluate this increase?

- Surface observations from the NOAA/ESRL flask network
- Nadir-mode retrievals from the GOSAT satellite
- Glint-mode retrievals from the GOSAT satellite

Turner *et al.*, GRL (2016)
NOAA/ESRL surface flask observations

Billings, OK (SGP)
Bermuda (BMW)

Increasing difference between continental US and background

Turner et al., GRL (2016)
What can we do with GOSAT?

- Can look at trends over locations where GOSAT observes
  - Many retrievals at coincident locations

- Do we find regional trend patterns?

Turner et al., GRL (2016)
Increasing difference in GOSAT trends

GOSAT and NOAA background are consistent

Contiguous US enhanced from background

Turner et al., GRL (2016)
Where do we find regional trends?

2010–2014 GOSAT trends in Δ methane

Doesn’t point to a particular source/sector

Turner et al., GRL (2016)
Observations of atmospheric methane and $\delta^{13}C$

-0.2‰ decrease in $\delta^{13}C$ during renewed growth

**Isotopic Composition ($\delta^{13}C$; ‰)**

- Pyrogenic = -22‰
- Thermogenic = -37‰
- Biogenic = -60‰

Schaefer *et al.* (2016):

Kirschke *et al.* (2013)
Need an increasing source with $\delta^{13}C = -59\%$.

Attribute decrease in $\delta^{13}C$ to biogenic sources.
Fossil fuel sources are not strictly thermogenic

“More than 20% of the world’s discovered gas reserves are of biogenic origin.”
Rice & Claypool, AAPG (1981)

“Non-conventional gas resources, such as coal-beds and organic-rich shales have largely been attributed to thermogenic processes, yet they may contain far more microbial gas than previously believed.”
Martini et al., Nature (1996)

“Shale-gas systems essentially are continuous-type biogenic (predominant), thermogenic, or combined biogenic-thermogenic gas accumulations.”
Curtis, AAPG (2002)
## Measured isotope compositions of methane sources

<table>
<thead>
<tr>
<th>Source</th>
<th>( \delta^{13}C )</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>fossil fuel (-15‰ to -76‰)</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gulf of Mexico</td>
<td>-58% to -60%</td>
<td>Stolper <em>et al.</em> (2014)</td>
</tr>
<tr>
<td>Antrim Shale</td>
<td>-50% to -53%</td>
<td>Stolper <em>et al.</em> (2015), Stolper <em>et al.</em> (2014)</td>
</tr>
<tr>
<td>Potiguar Basin</td>
<td>-37% to -50%</td>
<td>Stolper <em>et al.</em> (2014)</td>
</tr>
<tr>
<td>Haynesville Shale</td>
<td>-35% to -39%</td>
<td>Stolper <em>et al.</em> (2014)</td>
</tr>
<tr>
<td>Marcellus Shale</td>
<td>-34% to -36%</td>
<td>Stolper <em>et al.</em> (2014)</td>
</tr>
<tr>
<td>North Appalachia Basin</td>
<td>-26% to -36%</td>
<td>Wang <em>et al.</em> (2015)</td>
</tr>
<tr>
<td>Coal-associated gas</td>
<td>-15% to -70%</td>
<td>Zazzeri <em>et al.</em> (2016), Bréas <em>et al.</em> (2001)</td>
</tr>
<tr>
<td>Oil-associated gas</td>
<td>-30% to -60%</td>
<td>Bréas <em>et al.</em> (2001)</td>
</tr>
<tr>
<td>Gas drilling, pipeline leakage</td>
<td>-41% to -76%</td>
<td>Bréas <em>et al.</em> (2001)</td>
</tr>
<tr>
<td>Biomass burning</td>
<td>-17% to -32%</td>
<td>Dlugokencky <em>et al.</em> (2011), Bréas <em>et al.</em> (2001)</td>
</tr>
<tr>
<td><strong>non-fossil (-31‰ to -93‰)</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hydrates</td>
<td>-62% to -69%</td>
<td>Wang <em>et al.</em> (2015), Bréas <em>et al.</em> (2001)</td>
</tr>
<tr>
<td>Ruminants (C3 diet)</td>
<td>-61% to -76%</td>
<td>Bréas <em>et al.</em> (2001)</td>
</tr>
<tr>
<td>Ruminants (C4 diet)</td>
<td>-47% to -55%</td>
<td>Wang <em>et al.</em> (2015), Bréas <em>et al.</em> (2001)</td>
</tr>
<tr>
<td>Rice paddies</td>
<td>-50% to -68%</td>
<td>Bréas <em>et al.</em> (2001)</td>
</tr>
<tr>
<td>Landfills</td>
<td>-52% to -63%</td>
<td>Bréas <em>et al.</em> (2001)</td>
</tr>
<tr>
<td>Termites</td>
<td>-44% to -93%</td>
<td>Bréas <em>et al.</em> (2001)</td>
</tr>
</tbody>
</table>

Large overlap between fossil and non-fossil sources!
Based on isotope data, fossil fuel sources could explain between 0% and 100% of the renewed growth.
What can we say about the renewed growth?
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Can use a simple 2-box model:

\[
\frac{\partial [X]_N(t)}{\partial t} = E_{X,N}(t) - k[X][OH]_N(t)[X]_N(t) + \frac{[X]_S(t) - [X]_N(t)}{\tau_{NS}}
\]

\[
\frac{\partial [X]_S(t)}{\partial t} = E_{X,S}(t) - k[X][OH]_S(t)[X]_S(t) + \frac{[X]_N(t) - [X]_S(t)}{\tau_{NS}}
\]
Inputs to the 2-box model

- CH₄ Emissions (Tg/yr)
  - Global
  - Northern Hemisphere
  - Southern Hemisphere

- δ¹³CH₄ Composition (%)
  - Northern Hemisphere
  - Southern Hemisphere

- [OH] anomaly (%)
  - Global
  - Northern Hemisphere
  - Southern Hemisphere

- CH₃CCl₃ Emissions (Gg/yr)
  - Northern Hemisphere
  - Southern Hemisphere
Simulation with the uniformed inputs

Data from NOAA/ESRL\textsuperscript{1,2,3}, U. Heidelberg\textsuperscript{2}, UCI\textsuperscript{2}, UW\textsuperscript{2}, & GAGE/AGAGE\textsuperscript{3}

\begin{align*}
1 &= \text{CH}_4, \\
2 &= \delta^{13}\text{CH}_4, \\
3 &= \text{CH}_3\text{CCl}_3
\end{align*}
Consider a system represented by:

\[ y = F(x) + \epsilon \]

We can infer the most probable solution using Bayes Theorem:

\[
P(x|y) \propto P(y|x)P(x)
\]

Solving with a stochastic optimization.
Non-linear inversion

- Covariance Matrix Adaptation Evolution Strategy (CMA-ES)
  - Stochastic, derivative-free, non-linear/non-convex, optimization
  - Evolutionary algorithm that is less sensitive to local minima
Simulation with the posterior

CH\textsubscript{4} (ppb)

CH\textsubscript{3}CCl\textsubscript{3} (ppt)

Northern Hemisphere
Southern Hemisphere
Observations

\delta^{13}CH\textsubscript{4} (%)
Global

Northern Hemisphere

Southern Hemisphere

Posterior Emissions

Δ CH₄ Emissions (Tg/yr)

δ¹³CH₄ Composition (%)
Montzka et al. and McNorton et al. derive similar OH anomalies

Patra et al. infer a NH/SH OH ratio of $0.97 \pm 0.12$

Consistent with previous work looking at global mean OH
What if we don’t allow OH to vary?
What if we don’t allow OH to vary?

Biases can be aliased onto methane emissions.
What if we don’t allow CH$_4$ to vary?

Biases can be aliased onto OH anomalies
Summary

- Methane burden is sensitive to small changes
- Renewed growth is probably due to multiple factors
  - Changes in OH are a likely contributor
- Biases can be aliased from one species onto another

![Graph showing CH₄ (ppb) levels from 1985 to 2015 with marked periods of slowdown, plateau, and renewed growth. The data is courtesy of NOAA.]